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REPUBLIC OF SOUTH AFRICA
PATENTS ACT, 1978
APPLICATION FOR A PATENT AND
ACKNOWLEDGEMENT OF RECEIPT
(S. 30(1) Regulation 22)

REPUBLIC OF SOUTH AFRICA
FORM P.1 REVENUE
(to be lodged in duplicate)
-4.9.92 R 185.00
REFUBLIEX VAN SUID AFRIKA
A & A REF: 125136 AS/CST

THE GRANT OF A PATENT IS HEREBY REQUESTED BY THE UNDERMENTIONED APPLICANT
ON THE BASIS OF THE PRESENT APPLICATION FILED IN DUPLICATE

PATENT APPLICATION NO.	
21 01	926738
71	FULL NAMES(S) OF APPLICANT(S)

ISO-STER [PROPRIETARY] LIMITED

ADDRESS(ES) OF APPLICANT(S)

1 Waterpas Street, Isando, Extension 3, KAMPTON PARK, Transvaal Province, Republic of South Africa

54	TITLE OF INVENTION
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PROCESS FOR COATING A SUBSTRATE WITH A FILM OF THERMOPLASTIC POLYMERIC MATERIAL

Only the items marked with an "X" in the blocks below are applicable.

- ☒ THE APPLICANT CLAIMS PRIORITY AS SET OUT ON THE ACCOMPANYING FORM P.2. The earliest priority claimed is Country: ZA No: 91/6150 Date: 5 AUGUST 1991
- ☐ THE APPLICATION IS FOR A PATENT OF ADDITION TO PATENT APPLICATION NO. |21|01|
- ☐ THIS APPLICATION IS A FRESH APPLICATION IN TERMS OF SECTION 37 AND BASED ON APPLICATION NO. |21|01|

THIS APPLICATION IS ACCOMPANIED BY:

- ☒ A single copy of a provisional or two copies of a complete specification of 17 pages.
Drawings of 3 sheets.
- ☒ Publication particulars and abstract (Form P.8 in duplicate) (for complete only).
A copy of Figure of the drawings (if any) for the abstract (for complete only).
- ☒ An assignment of invention.
Certified priority document(s) (State quantity):
Translation of the priority document(s).
An assignment of priority rights.
- ☒ A copy of Form P.2 and the specification of RSA Patent Application No. |21|01| 91/6150
- ☒ A Form P.2 in duplicate.
- ☒ A declaration and power of attorney on Form P.3.
- ☐ Request for ante-dating on Form P.4.
- ☐ Request for classification on Form P.9.
- ☐ Request for delay of acceptance on Form P.4.

74 ADDRESS FOR SERVICE: *D.M. Kisch Inc. g/h*
~~Adams & Adams, Pretoria~~

DATED THIS 4TH DAY OF SEPTEMBER 1992

[Signature]
ADAMS & ADAMS
APPLICANTS PATENT ATTORNEYS

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A&A P201

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PRETORIA

FORM P7

REPUBLIC OF SOUTH AFRICA
Patents Act, 1978

COMPLETE SPECIFICATION

(Section 30(1) - Regulation 28)

OFFICIAL APPLICATION NO.

21 01

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LODGING DATE

22 4 September 1992

INTERNATIONAL CLASSIFICATION

51 B29C

FULL NAMES(S) OF APPLICANT(S)

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TITLE OF INVENTION

54 PROCESS FOR COATING A SUBSTRATE WITH A FILM OF THERMOPLASTIC
POLYMERIC MATERIAL

THIS INVENTION relates to a process for coating a substrate with a film of thermoplastic polymeric material and to a substrate having a thermoplastic polymeric coating film.

5 According to one aspect of the invention there is provided a process for coating a substrate, the process comprising forming a film of partially cross-linked thermoplastic polymeric material on at least part of the surface of the substrate. The process may include the step, prior to forming the film, of forming the partially cross-linked thermoplastic polymeric material. Forming
10 said partially cross-linked thermoplastic polymeric material may, further, include the step of partially cross-linking a thermoplastic polymeric starting material.

In other words, the process may comprise the steps of
15 subjecting a thermoplastic polymeric starting material which is capable of being cross-linked or optionally oxidized, to partial cross-linking, thereby to produce a thermoplastic partially cross-linked polymeric coating material and coating a substrate with the thermoplastic partially cross-linked polymeric coating material.

20 Subjecting the starting material to partial cross-linking may be by means of ionizing radiation or chemical cross-linking.

The thermoplastic polymeric starting material may be based on any suitable thermoplastic polymeric base material, which may be in the solid state or in solution or in emulsion form, and
25 which may be a plastics, resinous or elastomeric (rubbery) material, and may typically contain a suitable proportion of a

suitable cross-linking agent. Examples of base materials include homopolymers, co-polymers and ter-polymers of plastics materials and rubbers which are thermoplastic, and blends thereof, eg polyethylene, polypropylene, polyvinyl chloride, segmented co-polymers of butadiene and isoprene with styrene, natural rubber and polyethylene-ethylene propylene rubber blends.

Accordingly, the starting material may comprise a mixture of a thermoplastic polymeric base material and a cross-linking agent, the base material optionally being selected from the group consisting of: polyethylene, polypropylene, polyvinyl chloride, segmented co-polymers of butadiene and isoprene with styrene, natural rubber, blends of polyethylene, ethylene and propylene rubber, and mixtures of the foregoing, the mixture being in the form of a continuous phase of the base material in which the cross-linking agent is a discontinuous phase dispersed in finely divided form.

The method may include the preliminary steps of compounding or formulating the starting material, by admixing, with the polymeric base material, a suitable cross-linking agent. Thus, the process may include the step, prior to the step of partially cross-linking the thermoplastic polymeric starting material, of dispersing a cross-linking agent in the thermoplastic polymeric starting material. Usually the cross-linking agent will comprise at least two chemically reactive unsaturated bonds, such as double bonds, in its molecule, some of which unsaturated bonds react with the base material during the partial cross-linking under the influence of the radiation to produce the coating material. Suitable cross-linking agents include multifunctional acrylates or methacrylates such as trimethylolpropane trimethacrylate [TRIM] and allyl derivatives such as triallyl cyanurate [TAC]; and the material may be compounded to contain 0,5 - 25% by mass of cross-linking agent.

Compounding the material may take place in the usual way, eg by mixing the constituents to a sufficient degree of

homogeneity in the molten state at a temperature of typically not greater than 165°C and usually somewhat lower, and allowing the mixture to cool and set. Thus, the process may be one in which the thermoplastic polymeric starting material is solid, the dispersing of the cross-linking agent therein being by melting the thermoplastic polymeric starting material and cross-linking agent, mixing the cross-linking agent into the thermoplastic polymeric material while they are in molten form to form a mixture, and causing or allowing the mixture to solidify before it is partially cross-linked.

Accordingly, in a particular embodiment, the method may include the preliminary step, before the irradiation, of compounding or formulating the starting material by admixing a cross-linking agent into a molten polymeric thermoplastic base material, the cross-linking agent having one or more unsaturated bonds in its molecule, which bonds are capable of reacting with the base material under the influence of the irradiation to cross-link the base material, the starting material, if it is in a solid state, being allowed to set before the irradiation is carried out.

The step of forming the partially cross-linked thermoplastic polymeric material may include melting a thermoplastic polymeric starting material and dispersing a master batch of cross-linked material in the molten starting material. The process may thus include the step, prior to dispersing the master batch into the starting material, of forming the master batch by melting a thermoplastic polymeric diluent material and dispersing a cross-linking agent into the molten thermoplastic polymeric diluent material, the diluent material having a melting point at least 60°C lower than that of the starting material, and substantially fully cross-linking the diluent material.

The master batch may be caused or allowed to cool and set, the set master batch being subjected to a size reduction which renders it particulate before it is dispersed in the molten

starting material. The melting may be by heating to a temperature of at most 165°C. The dispersing of the cross-linking agent may, as mentioned above, be such as to form a mixture comprising the polymeric starting material which contains 0,5% - 25% by mass of the cross-linking agent.

As mentioned above, the process of forming the partially cross-linked material may include irradiating with ionizing radiation.

The irradiation process may be carried out according to the method described in the Applicant's South African Patent No. 89/6852. The partial cross-linking may thus be effected by subjecting the thermoplastic polymeric starting material to ionising radiation, and may be effected at ambient temperature. The process of irradiating may be by irradiating with radiation selected from α -, β - or γ -radiation. Thus radiation can be produced by a suitable radioactive isotope such as cobalt 60 or a suitable electron beam accelerator which generates energetic electrons with an energy of eg 50 keV to 10 MeV. The irradiating may be such as to cause the material being irradiated to absorb a radiation dose of 4 - 150 kGy. Thus, the absorbed radiation dose applied to said material during the partial cross-linking may be of the order of 4-150 kGy, and conventional electron beam machines or irradiators such as gamma irradiators can be employed for this purpose. In particular, the irradiation to which the starting material is subjected may be carried out at ambient temperature with the starting material in solid (or solution or emulsion) form by subjecting the starting material to an absorbed radiation dose to the starting material of 4-150 kGy.

As indicated above, in preferred embodiments of the invention, the cross-linking agent may be a member of the group consisting in multifunctional acrylates and methacrylates, multifunctional allyl derivatives and mixtures thereof and the irradiation cross-linking may be in accordance with the Applicant's South African Patent No. 89/6852 as mentioned above.

The nature of the base polymer and the cross-linking agent, the proportion of cross-linking agent and the degree of partial cross-linking by irradiation, will be selected so that these parameters produce a product with the desired physical and chemical properties. As at least a rough guide, known base polymer/cross-linking agent formulations can be employed which, when fully cross-linked by radiation, provide a product with desired physical and chemical properties.

The method may include, in addition to mixing the cross-linking agent into the base polymer, admixing one or more additives therein. Such additives include:

antioxidants [stabilizers] such as hindered phenol octadecyl 3-(3,5-di-t-butyl-4-hydroxyphenyl)propionate;

ultra-violet radiation stabilizers such as poly-([6-[(1,1,3,3-tetramethyl butyl)-imino]-1,3,5-triazine-2,4-diyl][2-(2,2,6,6-tetramethyl piperidyl)-imino]-hexamethylene-[4-(2,2,6,6-tetramethyl piperidyl)-imino]);

plasticizers such as phthalic esters or epoxidized soya oil;

impact modifiers such as ethylene-vinyl acetate copolymer or nitrile rubber;

slipping agents such as oleamide;

fillers such as calcium carbonate;

reinforcers such as carbon black;

flame retardants such as antimony trioxide and organobromine;

coupling agents such as silanes;

fungicides such as phosphites;

anti-blocking agents such as amide waxes;

pigments such as rutile;

essences such as dimethyl phenyl carbonyl;

lubricants such as stearic acid or calcium stearate;

blowing agents such as azodicarbonamide; and/or

electronic conductors such as carbon black.

The additives used and the proportions thereof used will be similar to those employed for similar purposes in polymeric

materials.

The irradiating may be carried out in an oxidizing atmosphere typically comprising 20 - 100% by volume oxygen.

5 The degree of cross-linking effected during the partial cross-linking by irradiation will be selected to provide a coating material which is thermoplastic; and the degree of cross-linking effected by the irradiation is typically sufficient to bind 1 - 50% by mass, eg 2 - 10% by mass, of the material, as determined by Soxhlet solvent extraction. Thus, forming the
10 partially cross-linked thermoplastic polymeric material may be such as to obtain a partially cross-linked polymeric material of which 1 - 50% by mass is cross-linked.

The coating process may be carried out according to any suitable coating process known in the art. Thus forming the film
15 may be by a coating process selected from:

flame spraying,
electrostatic powder coating,
powder dip-coating,
plasma dip-coating,
20 plasma spraying,
dipping from a hot melt, solution or emulsion,
fluidised bed methods and
electrodeposition.

25 An important feature of the invention arises from the fact that said partial cross-linking of thermoplastic polymers can lead to a marked enhancement in the adhesive properties of the cross-linked polymers to certain substrates, ie the partially cross-linked polymeric coating material adheres more strongly to the substrate than the base material from which it is made.

30 The adhesive nature of the thermoplastic polymer coating material can be influenced by the type and grade of the base material; the absorbed radiation dose and the dose rate during the irradiation; the atmosphere in which the irradiation of the

polymer is carried out; the nature of the substrate employed; the nature of the substrate surface; the temperature of the substrate and/or the coating material during the coating process; the contact time of the pre-irradiated polymer at said temperature; the cooling rate of the substrate following the coating thereof; and the coating process employed.

Thus, for example, the presence of oxygen in the atmosphere in which the irradiation takes place has in certain cases been found to be beneficial, in proportions of 20% by volume or more, up to 100% oxygen, air being conveniently employed. A rough substrate surface, eg shot-blasted or sand-blasted can be beneficial. Accordingly, routine experimentation should be employed with a view to optimizing the various parameters mentioned above, bearing practical and economic considerations in mind, so as to obtain an optimum or at least adequate product.

Substrates which can be used include wood, metals such as steel, glass, cardboard, paper, cement, concrete, screeding, plaster, tiles or brickwork, other polymeric materials such as plastics, rubbery or resinous materials, or the like. When the partially cross-linked thermoplastic polymeric coating material is used in solid form, it may be coated at a temperature above its melting point, with the substrate also heated above the melting point of the coating material. When the coating material is in the form of a solution or emulsion, the coating will typically take place at ambient temperatures.

As a development of the invention, the Applicant has found that forming a film coating in accordance with the process of the present invention for two substrates, the film being sandwiched therebetween can act adhesively to secure the substrates together. In accordance with this aspect of the invention, part of each substrate is coated, as described above, with the thermoplastic partially cross-linked polymeric coating material, the coated parts of the substrates being held together until the coating secures the substrates together. In other words the

process may be one in which forming the film of partially cross-linked thermoplastic polymeric material is on part of the surface of each of at least two substrates so that it is sandwiched between the two substrates, and acts adhesively to secure the substrates together. The process may further comprise the step, prior to forming the film, of treating the surface of each substrate to provide the substrate with a roughened surface.

The Applicant has accordingly, bearing in mind that the substrates can themselves be polymeric plastics, resinous or rubber materials, found that this aspect of the invention displays promise in joining plastics pipe lengths together spigot/socket fashion, the coating material in particulate or powder form being located in the spigot/socket joint where the pipe lengths nest together, and being melted to coat the nesting parts of the pipe lengths so that, when the melt is allowed to cool and solidify, the film or coating acts as an adhesive to secure the pipe lengths together. In this case the parts of the pipe lengths which nest together can be heated and dipped in a powder of the coating material, the powder forming a molten coating on the contact surfaces of the pipe lengths where they form the spigot/socket joint. The spigot will then be urged into the socket with the coating material molten, the coating material then cooling and solidifying, to join the pipe lengths sealingly together in adhesive fashion.

The invention will now be described, by way of example, with reference to the following Examples, giving details of tests carried out by the Applicant to demonstrate the invention, and with reference to the accompanying schematic drawings, in which:

Figure 1 shows an exploded side elevation of a spigot/socket joint between two pipe lengths to which the present invention is applicable;

Figure 2 shows a sectional side elevation of the joint of Figure 1;

Figure 3 shows an exploded sectional plan view of another spigot/socket joint between two pipe lengths to which the present

invention is applicable;

Figure 4 shows an exploded sectional plan view of a further spigot/socket joint between two pipe lengths to which the present invention is applicable;

5 Figure 5 shows a side elevation of the socket of the joint of Figure 4; and

Figure 6 shows an end elevation of the socket of Figure 5.

EXAMPLE 1

10 Linear low density polyethylene was subjected to gamma irradiation at various absorbed doses. Identical substrates which were panels made of polished tin-coated steel were coated by placing powder with a particle size of 600 μm thereon and heating to a temperature of 200°C to form a molten layer of polyethylene on the substrates. The irradiation was carried out under air or oxygen atmosphere and, after the coating step, cooling was effected at a rapid rate by quenching in water at 20°C/min or a slow rate of 12°C/min to ambient temperature. After cooling, 15 adhesion of the cooled polyethylene layer found on the substrates was tested. In each case the irradiation was carried out at a dose rate of 1,38 Gy/sec. Results are set forth in the following table, Table 1.

TABLE 1

Absorbed Dose (kGy)	Atmosphere	Adhesion Strength (N/mm)	Cooling Rate
0 (control)	-	0	Slow
8	Air	0	Slow
12	Air	0	Slow
16	Air	1,7	Rapid
16	Air	2,8	Slow
25	Air	2,1	Slow
25	Oxygen	3,5	Slow
33	Air	1,7	Slow

From Table 1 it is apparent that, for slow cooling, and for irradiation in air, no adhesion took place until an absorbed dose of 16 kGy had been dosed to the polyethylene. Rapid cooling at 16 kGy lead to a drop in adhesion strength and irradiation in oxygen at an absorbed does of 25 kGy led to an increase in adhesion strength. From these results it appears that adhesion can initially be achieved at a dose of between 12 and 16 kGy, and that the maximum adhesion strength is obtained at a dose between 12 and 25 kGy. It is to be emphasised that these results are subject to optimization, but it appears that there is a degree of partial cross-linking where adhesion strength is at a maximum, and that slow cooling and high proportions of oxygen in the irradiation atmosphere promote high adhesion strength.

EXAMPLE 2

Example 1 was repeated at a constant absorbed dose of 30 kGy with irradiation under oxygen. Results are set forth in table 2.

TABLE 2

Substrate	Adhesion Strength (N/mm)	Cooling Rate
Aluminium	0,6	Slow
Aluminium	0	Rapid
Tool Steel	6,5	Slow
Tool Steel	3,9	Rapid
Mild Steel	5,9	Slow
Mild Steel	5,3	Rapid

The results set forth in Table 2 confirm that slow cooling promotes high adhesion strength; and these results indicate that the nature of the substrate is important to adhesion strength, possibly because of differences in surface smoothness/roughness. In this regard it is to be noted that ancillary tests conducted by the Applicant have confirmed that

adhesion strength increases with increasing surface roughness of the substrate and decreases with increasing smoothness of the substrate.

5 From these tests it is clear that, for each substrate/coating material combination, routine tests will be required to optimize the absorbed dose. In general it appears that irradiation should take place in oxygen and that the substrate surface should be as rough as feasible while the cooling rate should be as slow as feasible. It also appears that adhesion strength increases with absorbed dose (which corresponds to the degree of partial cross-linking in the absence of oxygen), up to a maximum, after which adhesion strength decreases with further increased absorbed dose and cross-linking.

10 Turning to the drawings, Figures 1 and 2 show a spigot/socket joint 10 comprising two pipe lengths 12, 14, the length 12 having a socket formation 16 at one end thereof, which receives a spigot formation 18 at one end of the length 14, in nesting fashion. In the joint 10 the spigot formation 18 has a cylindrical radially outwardly facing contact surface 20, which nests in a corresponding cylindrical radially inwardly facing contact surface 22 on the socket formation 16.

25 In accordance with the invention powder particles of partially cross-linked polymeric coating material are melted on said contact surfaces 20,22 to coat them, and the spigot is thrust into the socket, followed by cooling to solidify the melt. The Applicant has found that, in this fashion, a sealed joint can be obtained from the pipe lengths in question, so that a pipeline of constant diameter can in principle be assembled from a plurality of lengths similar to lengths 12 and 14 shown in Figures 1 and 2, each having a spigot formation 18 at one end and a socket formation 16 at the other end, or each having two said spigot formations 18 or two said socket formations 16.

In Figure 3 a joint, generally designated 24, is shown, comprising two pipe lengths 14, each having a spigot formation 18 and, unless otherwise specified, the same reference numerals refer to the same parts as in Figures 1 and 2. Instead of a pipe length 12 with a socket formation 16 as shown in Figure 1, the joint 24 of Figure 3 employs a separate socket member 26 having a contact surface 28 and a pair of ears 30 for fastening it to a wall or the like support surface with nails, screws or the like. In this case the melt is formed on the surfaces 20 and 28 and the pipe lengths 14 are joined to the socket member 28 in essentially similar fashion to that described with reference to Figures 1 and 2 for the pipe lengths 12, 14.

In Figure 4 the same reference numerals refer to the same parts as in Figure 3 unless otherwise specified, the main differences between the joints of Figures 4 and 3 being that the socket 24 of Figure 4 employs pipe lengths 32 of constant diameter with no spigot formations, their contact surfaces 20 being provided by end zones thereof at 34; and that the socket member 26, instead of being thin-walled as in Figure 3 to provide a pipe of constant outer diameter, has a thickness the same as that of the pipe lengths 32 and is relatively thick-walled.

An optional stop is provided in the form of an internal circumferential rib 32 in the member 24 for the end zones (dotted lines).

Figures 5 and 6 show the socket member 26 of Figure 4 and once again, the same reference numerals refer to the same parts in Figures 5 and 6 as in Figures 3 and 4.

It is an advantage of the invention that it provides a process for permitting the coating of substrates with thermoplastic polymeric materials, and indeed, as demonstrated by the Examples, permits coating of certain substrates with acceptable adhesion strength, which substrates are not capable in similar circumstances of being coated by such polymeric

materials in their un-cross-linked state with any material adhesion strength whatsoever. It should also be noted that, after reaching a maximum, adhesion strength appears to reduce with further increases in absorbed irradiation dose. Furthermore, with reference to the drawings, the invention provides an elegant and easily applied method of assembling pipelines from pipe lengths.

CLAIMS

1. A process for coating a substrate, the process comprising forming a film of partially cross-linked thermoplastic polymeric material on at least part of the surface of the substrate.

5 2. A process as claimed in Claim 1, which includes the step, prior to forming the film, of forming the partially cross-linked thermoplastic polymeric material.

10 3. A process as claimed in Claim 2, in which forming the partially cross-linked thermoplastic polymeric material includes the step of partially cross-linking a thermoplastic polymeric starting material.

15 4. A process as claimed in Claim 3, which includes the step, prior to the step of partially cross-linking the thermoplastic polymeric starting material, of dispersing a cross-linking agent in the thermoplastic polymeric starting material.

20 5. A process as claimed in Claim 4, in which the thermoplastic polymeric starting material is solid, the dispersing of the cross-linking agent therein being by melting the thermoplastic polymeric starting material and cross-linking agent, mixing the cross-linking agent into the thermoplastic material while they are in molten form to form a mixture, and causing or allowing the mixture to solidify before it is partially cross-linked.

25 6. A process as claimed in Claim 2, in which forming the partially cross-linked thermoplastic polymeric material includes melting a thermoplastic polymeric starting material and dispersing a master batch of cross-linked material in the molten starting material.

30 7. A process as claimed in Claim 6, which includes the step, prior to dispersing the master batch into the starting material, of forming the master batch by melting a thermoplastic polymeric

diluent material and dispersing a cross-linking agent into the molten thermoplastic polymeric diluent material, the diluent material having a melting point at least 60°C lower than that of the starting material, and substantially fully cross-linking the diluent material.

8. A process as claimed in Claim 7, in which the master batch is caused or allowed to cool and set, the set master batch being subjected to a size reduction which renders it particulate, before it is dispersed in the molten starting material.

9. A process as claimed in any one of Claims 5 to 8 inclusive, in which the melting is by heating to a temperature of at most 165°C.

10. A process as claimed in any one of Claims 4 to 9 inclusive, in which the dispersing of the cross-linking agent is such to form a mixture comprising the polymeric starting material which contains 0,5 - 25% by mass of the cross-linking agent.

11. A process as claimed in any one of Claims 2 to 10 inclusive, in which forming the partially cross-linked material includes irradiating with ionizing radiation.

12. A process as claimed in Claim 11, in which the irradiating is by irradiating with radiation selected from α -, β - or γ -radiation.

13. A process as claimed in Claim 11 or Claim 12, in which the irradiating is such as to cause the material being irradiated to absorb a radiation dose of 4 - 150 kGy.

14. A process as claimed in any one of Claims 11 to 13 inclusive, in which the irradiating is carried out in an oxidizing atmosphere.

15. A process as claimed in Claim 14, in which the irradiating

is carried out in an oxygen-containing atmosphere comprising 20 - 100% by volume oxygen.

16. A process as claimed in any one of Claims 2 to 15 inclusive, in which forming the partially cross-linked thermoplastic polymeric material is such as to obtain a partially cross-linked polymeric material of which 1 - 50% by mass is cross-linked.

17. A process as claimed in any one of the preceding claims, in which forming the film is by a coating process selected from:

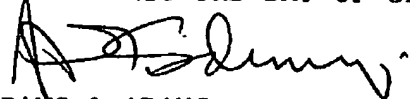
flame spraying,
electrostatic powder coating,
powder dip-coating,
plasma spraying,
dipping from a hot melt, solution or emulsion,
fluidised bed methods and
electrodeposition.

18. A process as claimed in any one of the preceding claims, in which forming the film of partially cross-linked thermoplastic polymeric material is on part of the surface of each of at least two substrates so that it is sandwiched between the two substrates, and acts adhesively to secure the substrates together.

19. A process as claimed in any one of the preceding claims, which comprises the step, prior to forming the film, of treating the surface of each substrate to provide the substrate with a roughened surface.

20. A process for coating a substrate, substantially as described and as illustrated herein.

DATED THIS 3RD DAY OF SEPTEMBER 1992


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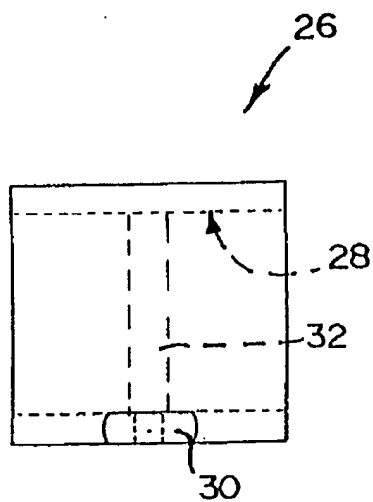


FIG 5

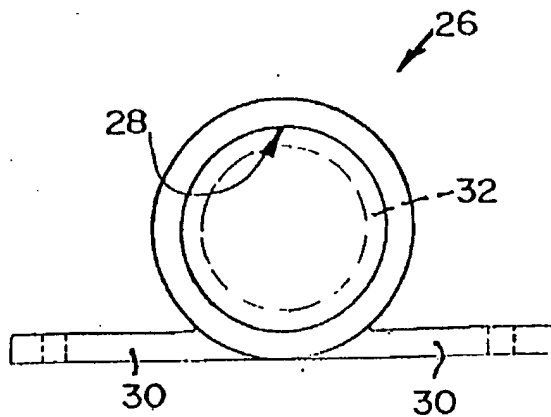



FIG 6


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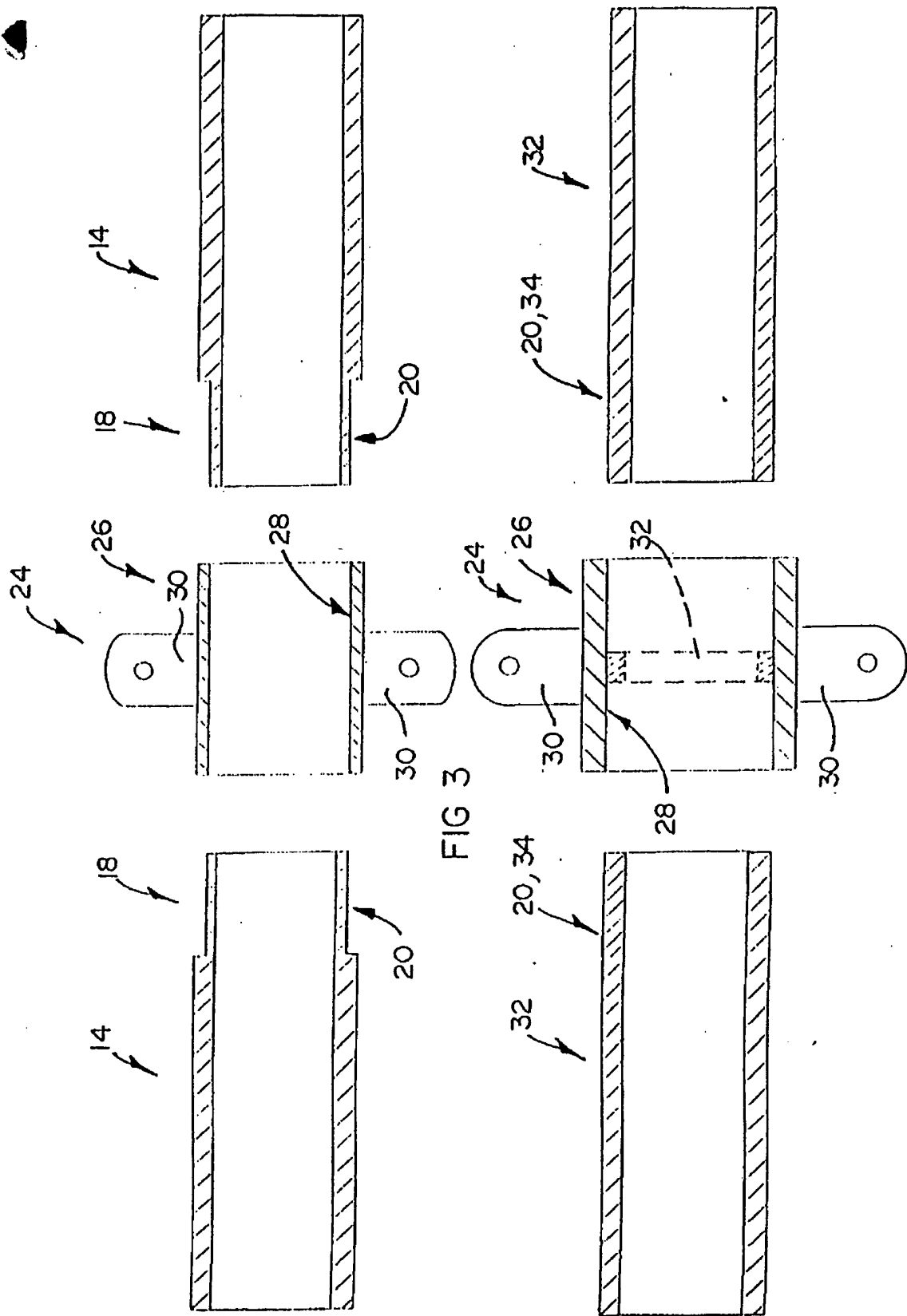
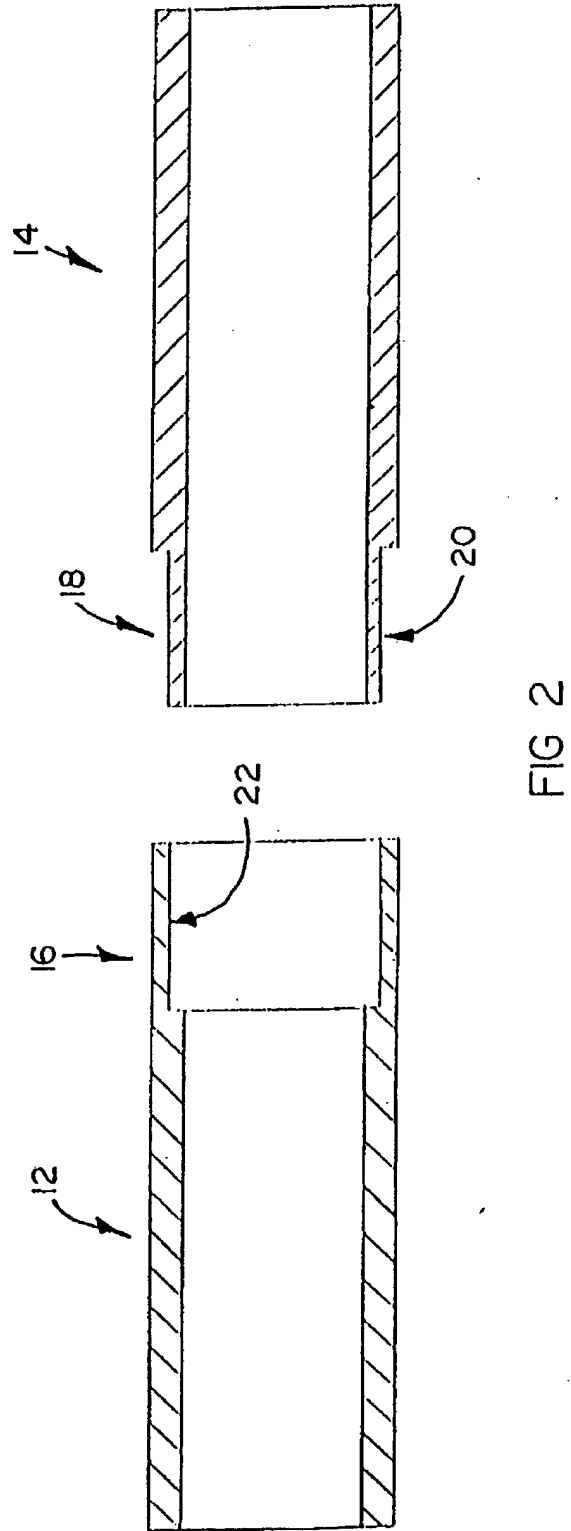
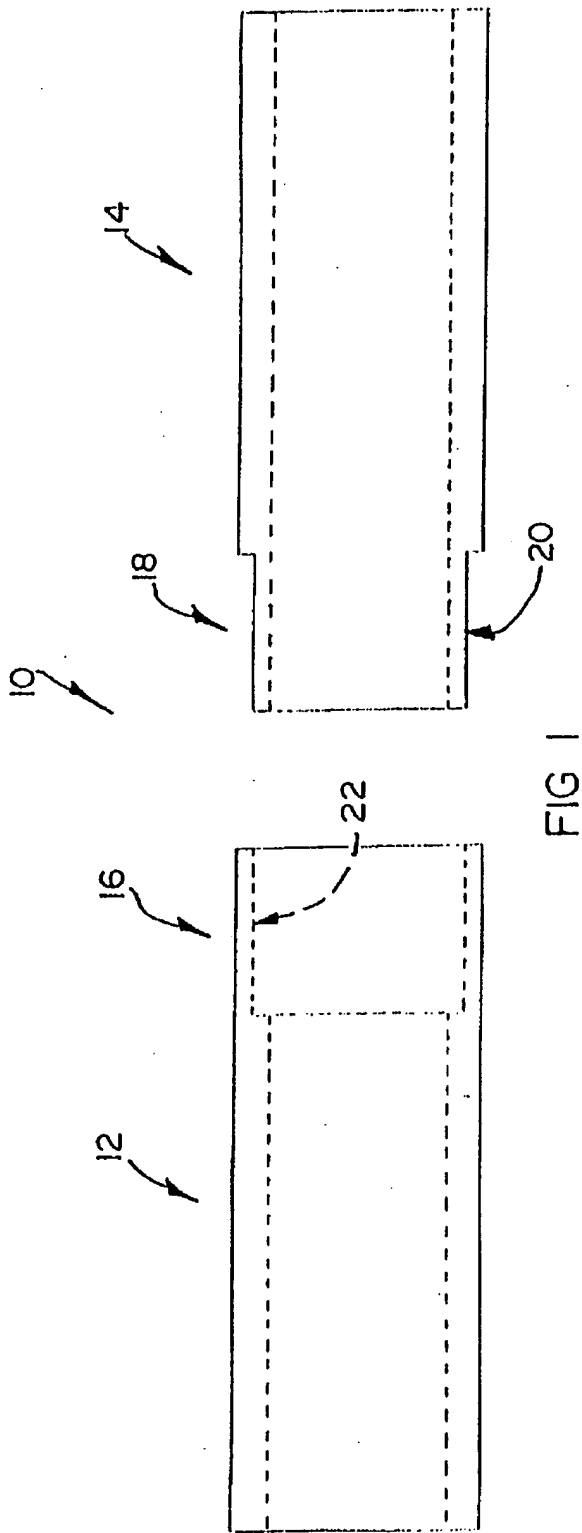


FIG 3

FIG 4

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